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PROCESS FOR PRODUCING MODIFIED OLEFIN POLYMER
[Kaishitsu Orefin Jugotai No Seizoho]

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TITLE (54) : PROCESS FOR PRODUCING MODIFIED
OLEFIN POLYMER

FOREIGN TITLE [54A] : Kaishitsu Orefin Jugotai No
Seizoho

SPECIFICATION

/1*

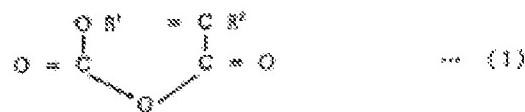
1. Title

Process for Producing Modified Olefin Polymer

2. Claims

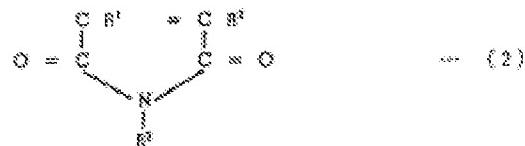
(1) A process for producing modified olefin polymers, said process comprising: bringing an olefin polymer in the form of particles into contact with an unsaturated carboxylic acid or a derivative thereof and an unsaturated silane compound in an inert gas in the substantial absence of a liquid medium and graft-copolymerizing them at a temperature lower than the melting temperature of said olefin polymer, using a radical initiator.

(2) The process stated in Claim 1, wherein the unsaturated carboxylic acid or its derivative is a minimum of one kind of compound selected from a group consisting of the compounds expressed by the general formula below:



and

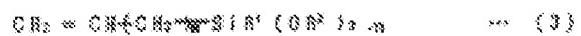
the compounds expressed by the general formula below:



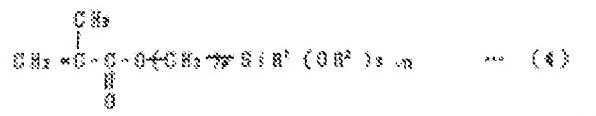
*Number in the margin indicates pagination in the foreign text.

(in the above general formulas, R¹ and R² represent hydrogen atoms or aliphatic hydrocarbon groups, and R³ represents an aliphatic hydrocarbon group or aromatic hydrocarbon group.)

(3) The process stated in Claim 1, wherein the unsaturated silane compound is a minimum of one compound selected from a group consisting of the compounds expressed by the general formula below:

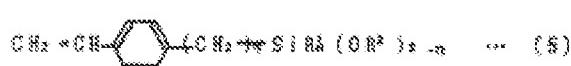


the compounds expressed by the general formula below:



, and

the compounds expressed by the general formula below:



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(in the above general formulas, R¹ and R² represent saturated aliphatic hydrocarbon groups and m represents 0, 1, 2, 3, or 4; n, 0, 1, or 2; p, 1, 2, 3, 4, or 5; and q, 0, 1, 2, 3, or 4.)

(4) The process stated in Claim 1, wherein the olefin polymer is homopolypropylene and a copolymer of propylene and an olefin other than propylene.

3. Detailed Description of the Invention

[Technical Field]

The present invention relates to a process for producing modified olefin polymers having excellent adhesion to metals, glass, inorganic fillers, and polar resins, such as polyvinyl alcohol and the like.

[Prior Art and Problems Thereof]

Olefin polymers have excellent mechanical properties, transparency, formability, and the like, but, on the other hand, they have the disadvantage of having poor adhesion to metals, glass, inorganic fillers, and polar resins because they are nonpolar. To ameliorate this shortcoming, there have been proposed methods of modifying olefin polymers with unsaturated carboxylic acids, anhydrides thereof, or esters thereof, such as acrylic acid, maleic acid, maleic anhydride, methyl acrylate, and the like, or with silane coupling agents, such as vinyl trimethoxysilane and the like.

The modified olefin polymers obtained by these methods have improved adhesion to metals, glass, inorganic fillers, and polar resins, but, because the decomposition reaction of the olefin polymers also takes place simultaneously in the modification process, it leads to various problems, such as difficulty in forming due to increased melt flow rates, impact resistance deterioration, and so forth.

Accordingly, for the purpose of preventing the decomposition of olefin polymers, in JP-A-S50-119888 is proposed a production method according to which, using a specific hydroperoxide as the graft initiator, maleic anhydride is graft-polymerized with polyolefin in a molten state at a temperature of from 120 °C to 250 °C, but, when polypropylene is used as the polyolefin, the melt flow rate increases, and the decomposition of polypropylene is not prevented well. As the method for modifying olefin polymers with unsaturated carboxylic acids,

anhydrides thereof, or esters thereof or with unsaturated silane compounds, such as vinyl trimethoxysilane and the like, the following two methods are known: the method of heating an olefin polymer at a temperature higher than its melting point and graft-polymerizing it in the presence of a radical initiator and the method of dissolving an olefin polymer in a solvent and subsequently graft-polymerizing it in the presence of a radical initiator. These methods, however, require a granulation or pulverization process later.

Olefin polymers, however, are often used in a powder or granular form, as in the case of preparing composite materials by mixing olefin polymers with powders of inorganic or organic substances, such as glass, calcium carbonate, carbon, talc, wood flour, and so forth, and, since the aforesaid methods do not produce polymers in a powder form, they are highly disadvantageous from the viewpoint of industrial production.

Besides the aforesaid two methods, there is, as the method of graft-copolymerizing a particulate olefin polymer as is, a method of suspending an olefin polymer, an unsaturated carboxylic acid, anhydride thereof, or ester thereof, or an unsaturated silane compound, such as vinyl trimethoxysilane or the like, and a radical initiator in a solvent in which the olefin polymer does not dissolve and graft-polymerizing them. This method, however, has a problem in that the formation of a graft polymer is inhibited due to the chain transfer of the growing graft polymer radicals caused by the solvent.

In JP-A-S50-077493 is proposed a method that, in the substantial absence of a liquid medium, brings a particulate olefin polymer into contact with maleic anhydride and graft-polymerizes them at a temperature lower than the melting temperature of said olefin polymer, using a radical initiator, but this method has a problem in that the olefin polymer after the modification has an increased flow melt rate. /3

The present inventors conducted extensive research to solve the aforesaid problems and, as a result, found that the concomitant use of an unsaturated carboxylic acid or its derivative and an unsaturated silane compound as the graft-use monomer in the graft copolymerization of an olefin polymer makes it possible to prevent the decomposition of the olefin polymer. Based on this finding, the present invention was achieved.

[Means for Solving the Problems]

The present invention is an olefin polymer production method for further improving the adhesion of olefin polymers to metals, glass, inorganic fillers, and polar resins, and it is a method for producing modified olefin polymers that is characterized by bringing an olefin polymer in the form of particles into contact with an unsaturated carboxylic acid or a derivative thereof and an unsaturated silane compound in an inert gas in the substantial absence of a liquid medium and graft-copolymerizing them at a temperature lower than the melting temperature of said olefin polymer, using a radical initiator, thereby

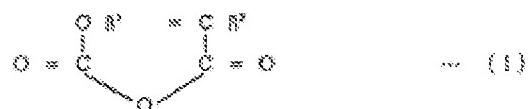
preventing the decomposition reaction of the olefin polymer in the modification process.

The olefin polymers that can be used in the present invention include: homopolymers, such as high pressure polyethylene, medium-low pressure polyethylene, polypropylene, and poly-1-butene; copolymers of these with other α -olefins; olefin polymer mixtures obtained by blending them with other olefin polymers; and so forth.

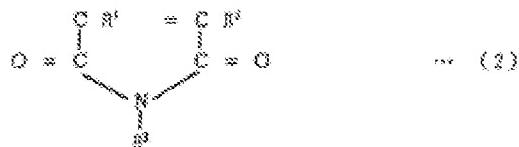
In the present invention, it is essential for these polymers to be in a state in which they are not melted or dissolved in a solvent, that is, for the form of these polymers to be a granular or powder form. It is particularly desirable for the polymer particles to have a diameter of from 0.001 to 4 mm, better yet, from 0.05 to 2 mm.

The unsaturated carboxylic acid or derivative thereof used in the present invention includes acrylic acid, methacrylic acid, maleic acid, fumaric acid, itaconic acid, ethyl acrylate, methyl acrylate, acrylamide, ethyl methacrylate, methacrylamide, monoethyl maleate, diethyl maleate, monoamide maleate, diamide maleate, maleimide, N-phenylmaleimide, and the like.

Especially desirable are the compounds expressed by the general formula below:



and the compounds expressed by the general formula below:

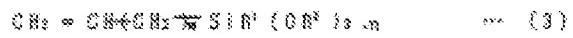


(in the above general formulas, R¹ and R² represent hydrogen atoms or aliphatic hydrocarbon groups, and R³ represents an aliphatic hydrocarbon group or aromatic hydrocarbon group.)

In more concrete terms, maleic anhydride, maleimide, N-phenyl maleimide, and the like are used. The proportion of the unsaturated carboxylic acid or derivative thereof to be used is typically from 0.1 to 50 % by weight, preferably from 1 to 20 % by weight, of the olefin polymer.

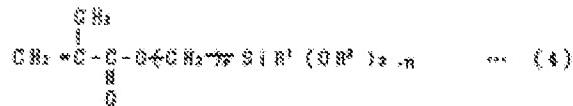
The unsaturated silane compound used in the present invention is a silane compound expressed by the general formula RSiR_nY_{4-n} (in which R represents an ethylenic unsaturated hydrocarbon group, alkoxy group, or aroyloxy [as transliterated] group; R, an aliphatic saturated hydrocarbon group; Y, a hydrolyzable organic group; and n, 0, 1, or 2.) In more concrete terms, the present invention can use the compounds in which R is a vinyl, allyl, isopropenyl, butenyl, cyclohexenyl, cyclopentadienyl, γ-methacryloxypropyl, styryl, β-styrylethyl, or the like group; R is a methyl, ethyl, propyl, decyl, or the like group; and Y is a methoxy, ethoxy, butoxy, formyloxy, acetoxy, propionyloxy, alkyl, arylamino, or the like group.

Especially desirable are the compounds represented by the general formula below:



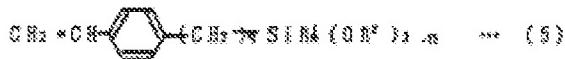
the compounds represented by the general formula below:

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, and

the compounds represented by the general formula below:



(in the above general formulas, R¹ and R² represent saturated aliphatic hydrocarbon groups and m represents 0, 1, 2, 3, or 4; n, 0, 1, or 2; p, 1, 2, 3, 4, or 5; and q, 0, 1, 2, 3, or 4.)

Concrete examples thereof include vinyl trimethoxysilane, allyl trimethoxysilane, γ-methacryloxypropyl trimethoxysilane, p-styryl trimethoxysilane, β-styrylethyl trimethoxysilane, and the like. The proportion of the unsaturated silane compound to be used is typically from 0.1 to 50 % by weight, preferably from 2 to 30 % by weight, of the olefin polymer.

The present invention can use various kinds of peroxides or azo compounds as the radical initiator. For example, benzoyl peroxide, di-tertiary butyl peroxide, dicumyl peroxide, azobisisobutyronitrile, and the like can be used. The proportion of the radical initiator to be used is typically from 0.01 to 20 % by weight, preferably from 0.2 to 10 % by weight of the olefin polymer.

The reaction temperature may be any temperature that is lower than the temperature at which the olefin polymer melts, but a

relatively high temperature is desirable for the purpose of increasing the graft ratio. The reaction is carried out typically at from 50 to 170 °C, preferably from 70 to 150 °C.

The reaction is carried out in an inactive gas, such as nitrogen, argon, or the like, in the substantial absence of a liquid medium. However, because maleic anhydride, maleimide, or the like used in the present invention sublimates, crystals of maleic anhydride, maleimide, or the like sometimes deposit at the low-temperature sections attached to the reactor--for instance, at a pressure gauge, stirrer, and the like mounted at the upper part of the reactor--and prevent the reaction from continuing. In such a case, it is desirable to use a small amount of a solvent that becomes a gas at a relatively low temperature, such as benzene, toluene, acetone, or the like, and thereby to prevent the crystal formation of maleic anhydride, maleimide, or the like.

As for the addition sequence of the components used, there is no specific limitation as long as the particulate polyolefin polymer, the unsaturated carboxylic acid or derivative thereof, the unsaturated silane compound, and the radical initiator make contact simultaneously.

After the completion of the polymerization, it is desirable to wash the reaction system with a solvent so as to eliminate the unsaturated carboxylic acid or derivative thereof, unsaturated silane compound, and radical initiator that have not been reacted and the unnecessary polymer of the unsaturated carboxylic acid or derivative

thereof and the unsaturated silane compound. As this solvent, the present invention can use solvents that can dissolve the unsaturated carboxylic acid or derivative thereof, unsaturated silane compound, radical initiator, and the polymer of the unsaturated carboxylic acid or derivative thereof and the unsaturated silane compound, examples of such solvents including toluene, acetone, methyl ethyl ketone, tetrahydrofuran, and the like.

[Effects of the Invention]

The polyolefin compositions produced according to the present invention attain improvements on the adhesion of polyolefins to metals, such as iron, aluminum, and the like, glass, and polar resins, such as polyvinyl alcohol, nylon, saponified ethylene-vinyl acetate copolymer, and the like. Furthermore, the present invention has the advantage that the decomposition reaction of polyolefins that occurs along with the graft copolymerization in the conventional methods can be prevented; consequently, forming difficulty caused by a melt flow rate increase or impact resistance deterioration can be prevented.

[Working Examples]

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The following presents working examples and comparative examples. Melt flow rate (MFR) was measured in accordance with JIS K 7210, and Izod impact strength (with notches) in accordance with JIS K 7110. The content rate of the unsaturated carboxylic acid or derivative thereof contained in a modified olefin polymer was determined by infrared absorption spectroscopy.

The content rate of the olefin polymer in a modified olefin polymer was determined with fluorescence X-ray. The content rates of the unsaturated carboxylic acid or derivative thereof and unsaturated silane compound that graft-polymerized to the olefin polymer in a modified olefin polymer were determined as follows: by dissolving the modified olefin polymer in a solvent completely and subsequently depositing it in acetone, the unsaturated carboxylic acid or derivative thereof and unsaturated silane compound that did not graft-polymerize to the olefin polymer were dissolved in acetone completely, and the unsaturated carboxylic acid or derivative thereof in the acetone-insoluble matter was determined by infrared absorption spectroscopy and the unsaturated silane compound therein with fluorescence X-ray.

Working Example 1

In a 10 L-capacity reactor equipped with agitator blades having spiral-shape double ribbons, 500 g of a polypropylene powder (MFR = 30) was placed, and, after the reactor was thoroughly purged with a nitrogen gas, 80 g maleic anhydride, 10 g benzoyl peroxide, and 50 mL toluene were added. After the temperature inside the reaction system was raised to 90 °C, 90 g vinyl trimethoxysilane was injected into the system, and a reaction was carried out for 5 hours at 90 °C while the reaction system was agitated.

After the completion of the reaction, thorough washing with acetone and subsequent drying under reduced pressure were carried out.

The yield of the modified olefin polymer was 540 g, and its MFR was 1.0. The content rates of the maleic anhydride and vinyl trimethoxysilane in the modified olefin polymer were 2.7 % by weight and 4.3 % by weight.

The content rates of the maleic acid and vinyl trimethoxysilane that were grafted to the polypropylene in the modified olefin polymer were 1.6 % by weight and 2.1 % by weight.

Working Example 2

As the unsaturated carboxylic acid, 50 g N-phenyl maleimide was used, and, as the unsaturated silane compound, 70 g γ -methacryloxypropyl trimethoxysilane was used. The rest was the same as in Working Example 1. The yield of the modified olefin polymer was 550 g, and its MFR was 4.1. The content rates of the N-phenyl maleimide and γ -methacryloxypropyl trimethoxysilane in the modified olefin polymer were 3.2 % by weight and 6.0 % by weight.

The content rates of the N-phenyl maleimide and γ -methacryloxypropyl trimethoxysilane that were grafted to the polypropylene in the modified olefin polymer were 1.5 % by weight and 2.2 % by weight.

Working Example 3

As the unsaturated carboxylic acid, 50 g maleic anhydride was used; as the unsaturated silane compound, 70 g β -styrylethyl trimethoxysilane was used; and, as the radical initiator, 7 g dicumyl peroxide was used. The reaction temperature was set to 120 °C. The

rest was the same as in Working Example 1. The yield of the modified olefin polymer was 560 g, and its MFR was 2.4. The content rates of the maleic anhydride and β -styrylethyl trimethoxysilane in the modified olefin polymer were 3.1 % by weight and 7.3 % by weight.

The content rates of the maleic anhydride and β -styryl ethyl trimethoxysilane that were grafted to the polypropylene in the modified olefin polymer were 1.4 % by weight and 3.5 % by weight.

Comparative Example 1

As the solvent, 5000 mL toluene was used. The rest was the same as in Working Example 1. The yield of the modified olefin polymer was 510 g, and its MFR was 30. The content rates of the maleic anhydride and vinyl trimethoxysilane in the modified olefin polymer were 0.68 % by weight and 1.1 % by weight.

The content rates of the maleic anhydride and vinyl trimethoxysilane that were grafted to the polypropylene in the modified olefin polymer were 20 % by weight and 0.31 % by weight.

Comparative Example 2

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Polypropylene (MFR = 1.8) was used, and maleic anhydride was not used. The rest was the same as in Working Example 1. The yield of the modified olefin polymer was 520 g, and its MFR was 2.7. The content rate of vinyl trimethoxysilane in the modified olefin polymer was 3.8 % by weight.

The content rate of the vinyl trimethoxysilane that was grafted to the polypropylene in the modified olefin polymer was 1.7 % by weight.

Comparative Example 3

Polypropylene (MFR = 1.8) was used, and vinyl trimethoxysilane was not used. The rest was the same as in Working Example 1. The yield of the modified olefin polymer was 500 g, and its MFR was 5.3. The content rate of maleic anhydride in the modified olefin polymer was 0.16 % by weight.

The content rate of the maleic anhydride that was grafted to the polypropylene in the modified olefin polymer was 0.10 % by weight.

Working Example 4

With a Henschel mixer (a product name), 99.8 % by weight of the modified polyolefins obtained in Working Examples 1 through 3 were each mixed thoroughly with 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate. The mixtures thus obtained were formed into test pieces with an extruder that was set at 230 °C, and the Izod impact strengths of the test pieces were measured. The results are shown in Table 1.

Comparative Example 4

With a Henschel mixer (a product name), 99.8 % by weight of the modified polyolefins obtained in Comparative Examples 1 through 3 were each mixed thoroughly with 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate. The mixtures thus obtained were formed into test

pieces with an extruder that was set at 230 °C, and the Izod impact strengths of the test pieces were measured. The results are shown in Table 1.

Comparative Example 5

With a Henschel mixer (a product name), 99.8 % by weight of the polyolefins used in Working Examples 1 through 3 and Comparative Examples 1 through 3 were each mixed thoroughly with 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate. The mixtures thus obtained were formed into test pieces with an extruder that was set at 230 °C, and the Izod impact strengths of the test pieces were measured. The results are shown in Table 1.

Working Example 5

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the modified polyolefins obtained in Working Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR: 3.0, ethylene: 8 % by weight), and 0.1 [sic] each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. These pellets were fed to a 230 °C T-die film-forming apparatus, thereby preparing 500 μ -thick sheets.

Next, 300 μ -thick steel sheets (JIS G3141 SPCC-SB) were degreased with trichloroethylene. The obtained polyolefin sheets (A) and the steel plates (B) were arranged in three layers in the order of B-A-B, and, using a compression forming machine, the resulting structures were heated and pressed at an adhesion temperature of 220 °C under a

pressure of 20 atm for 1 minute and subsequently cooled, thus preparing composite plates. The T-peel strengths of these composite plates were determined in accordance with JIS 6854. The results are shown in Table 2.

Comparative Example 6

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the modified polyolefins obtained in Comparative Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR: 3.0, ethylene: 8 % by weight), and 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. The rest was conducted in the same manner as in Working Example 5. The results are shown in Table 2.

Comparative Example 7

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the polyolefins used in Working Examples 1 through 3 and Comparative Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR: 3.0, ethylene: 8 % by weight), and 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. The rest was conducted in the same manner as in Working Example 5. The results are shown in Table 2.

Working Example 6

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the modified polyolefins obtained in Working Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR:

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3.0, ethylene: 8 % by weight), and 0.1 [sic] each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. These pellets were fed to a 230 °C T-die film-forming apparatus, thereby preparing 50 μ -thick films.

The obtained polyolefin films (A) and 20 μ -thick ethylene-vinyl acetate copolymer films (Kuraray Eval Film EF-F) (B) were arranged in three layers in the order of A-B-A, and, using a heat-seal apparatus, the resulting structures were heated and pressed at an adhesion temperature of 200 °C under a pressure of 1 atm for 1 second. The edges of the two layers--that is, the ethylene-vinyl acetate copolymer film and the modified polyolefin film--that made contact with the plane for measuring the adhesion peel strength of the thus-formed composite film were held with the chucks of an autograph manufactured by Shimazu Corporation, and the stress detected at a peel rate of 50 mm/minute was measured and taken as the peel strength. The results are shown in Table 2.

Comparative Example 8

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the modified polyolefins obtained in Comparative Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR: 3.0, ethylene: 8 % by weight), and 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. The rest was conducted in the same manner as in Working Example 6. The results are shown in Table 2.

Comparative Example 9

Using a Henschel mixer (a product name) and an extruder, 30 % by weight of the polyolefins used in Comparative [sic] Examples 1 through 3 and Comparative Examples 1 through 3, 69.8 % by weight of a propylene-ethylene copolymer (MFR: 3.0, ethylene: 8 % by weight), and 0.1 % by weight each of 2,6-di-butyl-cresol and calcium stearate were kneaded and formed into pellets. The rest was conducted in the same manner as in Working Example 6. The results are shown in Table 2.

TABLE 1

No	(a) オレフィン重合体組成物			M F R (g/10min)	アイゾット衝撃強度 (kg f · cm/cm ²) (f)
	オレフィン重合体 (b)	含有率(c) (重量%)	不飽和カルボン酸 誘導体(d) 化合物(e)		
1	改質オレフィン重合体 (実施例-1)	2.7	4.3	1.3	6.35
2	改質オレフィン重合体 (実施例-2)	3.2	6.0	4.3	4.03
3	改質オレフィン重合体 (実施例-3)	3.1	7.3	2.5	4.36
4	改質オレフィン重合体 (比較例-1)	0.68	1.1	3.3	2.10
5	改質オレフィン重合体 (比較例-2)	0	2.3	2.9	3.53
6	改質オレフィン重合体 (比較例-3)	0.16	0	5.3	3.01
7	ポリプロピレン (MFR 3.0)	0	0	3.1	2.41
8	ポリプロピレン (MFR 1.8)	0	0	2.0	4.17

Key: 1-6) modified olefin polymer; 7, 8) polypropylene; a) olefin polymer composition; b) olefin polymer; c) content rate; d) unsaturated carboxylic acid derivative; e) unsaturated silane compound; f) Izod impact strength.

TABLE 2

No.	オレフィン重合体 (b)	(a) オレフィン重合体組成物					結(イ)果	
		割 (c)	合 (重量%)	含 有 (f) 率 (重量%)	MFR	剥離強度(j) (kgf/25mm wide)	鋼 板 (k)	エバール (l)
	オレフィン重合体 (d)	オレフィン 重合体 (e)	不飽和 カルボン酸 誘導体 (g)	不飽和 シラン 化合物 (h)	(g)/(h)(n)			
1	実施例 - 1	30.0	69.8	0.81	1.29	2.4	38.7	1.38
2	実施例 - 2	30.0	69.8	0.96	1.80	3.3	25.1	1.95
3	実施例 - 3	30.0	69.8	0.93	2.19	3.0	30.9	1.66
4	比較例 - 1	30.0	69.8	0.20	0.33	6.4	7.0	0.41
5	比較例 - 2	30.0	69.8	0	0.69	3.1	4.4	0.19
6	比較例 - 3	30.0	69.8	0.048	0	3.7	3.6	0.20
7	ポリプロピレン (MFR 3.0)	30.0	69.8	0	0	6.2	接着せず (m)	接着せず (m)
8	" (MFR 1.8)	30.0	69.8	0	0	2.6	接着せず (m)	接着せず (m)

Key: 1-3) working example; 4-6) comparative example; 7) polypropylene;
 a) olefin polymer composition; b) olefin polymer; c) ratio; d) olefin polymer;
 e) propylene-ethylene acid [sic] copolymer; f) content rate;
 g) unsaturated carboxylic acid derivative; h) unsaturated silane
 compound; i) results; j) peel strength (kgf/25 mm wide); k) steel
 plate; l) Eval; m) did not bond.

WRITTEN AMENDMENT

September 30, 1988

To: Director-General of the Patent Office

1. Case Identification

S62 [1987] Patent Application No. 264,966

2. Title of the Invention

Method for producing modified olefin polymer

3. Party filing the amendment

Relation to the case: Patent applicant

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(Postal Code 530)

(207) Chisso Corporation.

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Shinjuku, Shinjuku-ku, Tokyo-to (Postal Code 160)

(6601) Patent Attorney, Yataro Sasai

(Tel: 354-1285)

5. Date of invitation to correct

Voluntary amendment

6. Number of inventions to be increased by the amendment

None

7. Parts amended

The claims section and the "detailed description of the invention" section in the specification

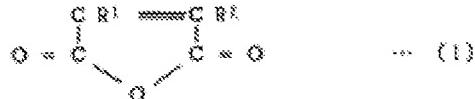
8. Details of the amendment

The specification shall be amended as follows.

I. The claims section in its entirety shall be amended as per attachment.

II. The "detailed description of the invention" section shall be amended as follows.

- (1) Amend "0.001 to 4 mm" in line 13 on page 8 to "0.001 to 10 mm."
- (2) Amend "0.05 to 5 mm" in the same line on the same page to "0.1 to 5 mm."
- (3) Amend the entire content of line 6 on page 9 to:



- (4) Insert "hydrogen," between " R^3 " and "aliphatic" on line 4 from the bottom of the same page.
- (5) Amend " $RSiR_nY_{n-n}$ " in line 5 on page 10 to " $R^1SiR^2_nY_{3-n}$ ".
- (6) Amend "here, R" in the same line on the same page to "here, R^1 ".
- (7) Amend "R" in line 7 on the same page to " R^2 ".
- (8) Amend "expressed" in line 9 on the same page to "expressed by."
- (9) Amend "R" in line 10 on the same page to " R^1 ".
- (10) Amend "R" in line 13 on the same page to " R^2 ".
- (11) Amend the entire content of the last line of the same page to:

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CH2=CH(C6H5)C(=O)S(=O)(=O)C6H5 ** (3)

(12) Amend the entire content of line 3 of page 11 to:

CH2=C(CH2C(=O)S(=O)(=O)C6H5)C6H5 ** (4)

(13) Amend "0.2 to 10 % by weight" in lines 8 to 9 on page 12 to "0.2 to 5 % by weight."

(14) Delete ", maleimide" from lines 4 and 3 from the bottom of the same page and from the last line of the same page.

(15) Delete ", maleimide" from line 4 of page 13.

(16) Delete "unsaturated" from line 5 of page 15.

(17) Amend "in the olefin polymer" in line 8 of the same page to "the silane compound."

(18) Delete "unsaturated" in lines 4 to 3 and line 2 from the bottom of the same page.

(19) Insert "a derivative of" between "As" and "the unsaturated carboxylic acid" in line 2 on page 17.

(20) Amend "20 % by weight" in line 2 from the bottom of page 18 to "0.2 % by weight."

(21) Amend "0.1" in line 10 on page 21 to "0.1 % by weight."

(22) Amend "0.1" in line 6 on page 23 to "0.1 % by weight."

(23) Amend "unsaturated carboxylic acid derivative" in Table 1 on page 25 to "carboxylic acid derivative."

- (24) Amend "unsaturated silane compound" in the same table on the same page to "silane compound."
- (25) Amend the MFR value "3.3" of No. 4 in the same table on the same page to "33."
- (26) Amend "unsaturated carboxylic acid derivative" and "unsaturated silane compound" in Table 2 on page 26 to "carboxylic acid derivative" and "silane compound", respectively.

9. List of attached documents

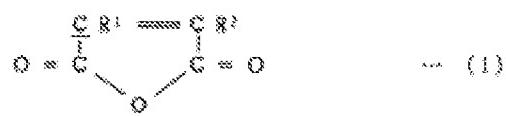
Attachment (the entire claims section)

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ATTACHMENT (the entire claims section)

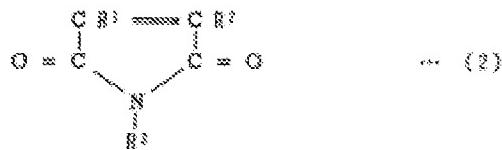
(1) A process for producing modified olefin polymers, said process comprising: bringing an olefin polymer in the form of particles into contact with an unsaturated carboxylic acid or a derivative thereof and an unsaturated silane compound in an inert gas in the substantial absence of a liquid medium and graft-copolymerizing them at a temperature lower than the melting temperature of said olefin polymer, using a radical initiator.

(2) The process stated in Claim 1, wherein the unsaturated carboxylic acid or its derivative is a minimum of one kind of compound selected from a group consisting of the compounds expressed by the general formula below:



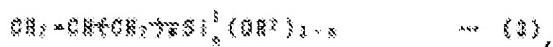
and

the compounds expressed by the general formula below:

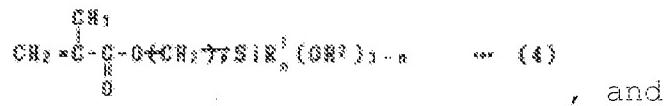


(in the above general formulas, R¹ and R² represent hydrogen atoms or aliphatic hydrocarbon groups, and R³ represents a hydrogen atom, an aliphatic hydrocarbon group, or aromatic hydrocarbon group.)

(3) The process stated in Claim 1, wherein the unsaturated silane compound is a minimum of one compound selected from a group consisting of the compounds expressed by the general formula below:



the compounds expressed by the general formula below:



the compounds expressed by the general formula below:



(in the above general formulas, R¹ and R² represent saturated aliphatic hydrocarbon groups and m represents 0, 1, 2, 3, or 4; n, 0, 1, or 2; p, 1, 2, 3, 4, or 5; and q, 0, 1, 2, 3, or 4.)

(4) The process stated in Claim 1, wherein the olefin polymer is homopolypropylene and a copolymer of propylene and an olefin other than propylene.